

Electrokinetic restoration of saline agricultural lands

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Received: 27 August 2009 / Accepted: 4 January 2010 / Published online: 14 January 2010
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Abstract Salinization of greenhouse soils has become a serious problem in Korea because of the extensive use of chemical fertilizers to improve crop yield. This study investigated the feasibility of electrokinetic (EK) treatment for reclamation of saline soil. Experiments were conducted using voltage gradients of 1, 2, and 3 V/cm applied for 48 and 96 h. Anions such as chloride, sulfate, and nitrate were transported toward the anode and accumulated there, whereas cations were transferred toward the cathode by electromigration. Among the various ions, the highest removal efficiency was achieved for nitrate: >80% at 48 h and >99% at 96 h. Chloride removal after 96 h was substantially higher than that after 48 h because the longer period of time allowed more electrical transport via electromigration and electro-osmosis. However, the removal efficiency for sulfate and calcium did not change significantly between 48 and 96 h. Soil EC was lower than the initial value in all soil sections at 96 h. The lowest value, 1.8 dS/m, was seen in the experiment employing a gradient of 3 V/cm for 48 h. This study demonstrated that nitrate can be readily removed from soil by electromigration. Further, other ions can also be removed by EK treatment; therefore, it could be successfully used for reclamation of saline soils.

Keywords Electrokinetic remediation · Voltage gradient · Electric conductivity · Saline soil · Salts

1 Introduction

Salinity has compromised approximately 7% of the earth's land, 20% of agricultural lands, and 50% of the cultivated land in the world [1–4]. In agricultural lands, the high salinity was mainly caused by overuse of fertilizers and chemical pesticides to improve the crop productivity, and insufficient irrigation [5–9]. The use of greenhouses has dramatically increased due to the increase in demand for greenhouse crops and because of the large yields that can be achieved [10]. Greenhouses in Korea account for 3.5% of the total agricultural area. Repeated cultivation is generally performed and manure is spread over the field. The closed system of a greenhouse is designed to create a particular environment in which a high inside temperature is maintained and rainfall is intercepted [11]. However, consequently, salts from the fertilizers do not percolate into the subsoil and are not washed away by the rain as they would in an open field. Furthermore, the high greenhouse temperature increases evaporation, causing salt accumulation at the soil surface. Moreover, the repeated cultivation accelerates soil salinization. Many species of salts exist in ionic form in the soil: cations include sodium, calcium, potassium, magnesium, etc.; anions include chloride, sulfate, nitrate, and phosphate, etc. A certain quantity of salts is essential for plant growth, and all soils contain some salts in pore water. However, an excess amount of salt in soil can have a detrimental effect on plant growth, including reduced water availability, osmotic adjustment, and nutrient uptake and also tissue injury [6]. Excess nitrate can have adverse effects not only on plants, but also on humans, e.g., methemoglobinemia (blue baby syndrome) and epidemiological gastric cancer [12, 13]. Further, surplus Na^+ causes the virulence to plants and degrades

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soil physical properties, and Cl^- can be toxic. The salinity of soil can be expressed in term of its electric conductivity (EC), which is measured in deci-Siemens per meter (dS/m). There does not appear to be any risk for plants in soils with EC below 2 dS/m. Generally, soil salinity in agricultural lands is reduced by irrigation, by mixing in earth from another place, or by exchanging the saline soil with clean soil in places where soil EC is excessive [5, 14].

These methods require large volumes of water and fresh soil. By contrast, electrokinetic (EK) remediation is a low-cost method that can be used on soil with poor permeability and can be applied in situ [15–19]. The EK process involves direct application of a low voltage gradient or electrical current to polluted soil through installed electrodes [20–23]. Salts, which have high water solubility and exist as charged ions, are transported and removed by electromigration (movement of charged ions), electro-osmosis (liquid flow from anode to cathode through the pore spaces between particles), and electrophoresis (transfer of charged particles) from the soil surface [20, 24]. Recently, several studies on desalination using EK remediation have been reported [7, 12–14, 24–27].

Budhu et al. [25], Eid et al. [7], and Jia et al. [27] evaluated nitrate movement using a lab-scale EK system and confirmed nitrate movement toward the anode by electromigration. Furthermore, they found that nitrate migration and accumulation was greater when the electric intensity was greater, the initial nitrate concentration was lower, and the electricity had been applied for a longer duration. The effects of nitrate movement by electromigration under a hydraulic gradient were reported by Jia et al. [13, 27]. In these laboratory-scale studies, vertical soil columns with the anode located at the top were used and nitrate solution was injected at the anode. In a similar study, Manokarajah and Ranjan [12, 26] used a horizontal column system. Their findings show that the nitrate accumulated at the anode by electromigration under EK is retained there even under hydraulic flow toward the cathode. Eid et al. [14] used numerical simulation to predict the nitrate concentration after EK treatment on the basis of soil pH and initial concentration. Furthermore, a field electroreclamation test using a horizontal drainage system was conducted by Cairo et al. [24].

There was no report on EK restoration of agricultural saline field soil. Therefore, laboratory-scale EK remediation was conducted on saline soil collected from a greenhouse in this study. The effects of voltage gradient and duration on the mobility and removal efficiency of salts were examined. In addition, the viability of EK remediation was evaluated to reduce the soil EC affected to plant growth.

2 Materials and methods

2.1 Saline soil

In the present study, soil was collected from vinyl greenhouses in which pumpkins and peppers were cultivated. The collected soil was air-dried and mixed uniformly. Next, this soil was sieved through a 2-mm screen and its water content was adjusted to approximately 30% with tap water prior to the EK experiment. Table 1 shows the chemical and physical properties of the soil; it was classified as silty loam.

2.2 EK experiment

Figure 1 shows a schematic of the EK remediation system. A soil compartments (4 cm × 4 cm × 20 cm) were placed between electrode chambers (4 cm × 4 cm × 4 cm) and were filled with compacted saline soil. Electrode compartments acted as overflow systems. Tap water was used

Table 1 Physico-chemical properties and composition of saline soil

Parameters	Value
pH	5.98
EC (dS/m)	4.5
Organic content (%)	5.18
CEC (meq/100 g)	35.72
Particle size distribution (%)	
Sand	35.72
Silt	56.18
Clay	8.1
Initial concentration (mg/kg)	
Cl^-	189.72
NO_3^-	2455.83
SO_4^{2-}	603.35
Ca^{2+}	1934.53
K^+	3085.33
Na^+	840.53

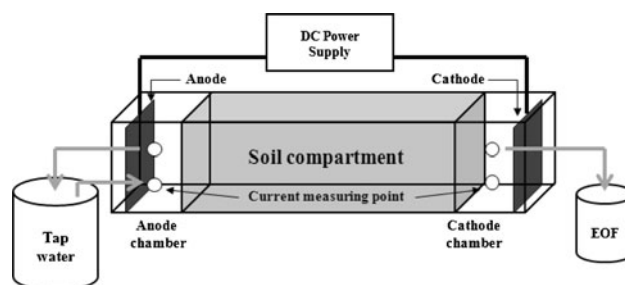


Fig. 1 Diagram for electrokinetic experiment

Table 2 Experimental design for electrokinetic process

Experiments	Voltage gradient (V/cm)	Duration (h)
48-1	1	48
48-2	2	48
48-3	3	48
96-1	1	96
96-2	2	96
96-3	3	96

as the electrolyte; it filled the electrode compartment and circulated through the anode compartment continuously. The overflowed electro-osmotic flow (EOF) at the catholyte reservoir was collected in a connected bottle. A graphite and titanium-coated platinum electrodes were used as cathode and anode, respectively.

In this study, six EK cells were set-up and subjected to a constant voltage gradient of 1, 2, and 3 V/cm. Three experiments were run for 48 h and the other three for 96 h (Table 2). During EK treatment, the electrical current was measured at the electrode compartment indicated in Fig. 1, and EOF volume measurement was also conducted.

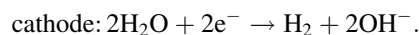
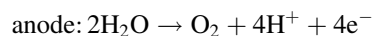
2.3 Analysis

Soil samples were analyzed to determine the removal efficiency and movement of salts. Soil EC and pH were measured with 10 g of powdered soil and 50 mL of distilled water using a conductivity meter (Istek 455C, Korea) and pH/ISE meter (Istek 735P, Korea), respectively. Anions such as Cl^- , NO_3^- , and SO_4^{2-} were extracted from soil samples mixed with distilled water in the ratio of 1:5. A 5-g sample of soil and 50 mL of 0.1 N NH_4OAc ($\text{CH}_3\text{COONH}_4$) adjusted to pH 7 using CH_3COOH and NH_4OH were mixed thoroughly in order to extract the exchangeable cations from soil [28]. These soil and solution mixtures were filtered and analyzed. Atomic adsorption spectrometry (AAS; AA 6300, Shimadzu, Japan) and ion chromatography (IC; Futecs, Korea) were used to determine concentration of cations and anions, respectively, in extracts and solutions such as the electrolyte and EOF [29].

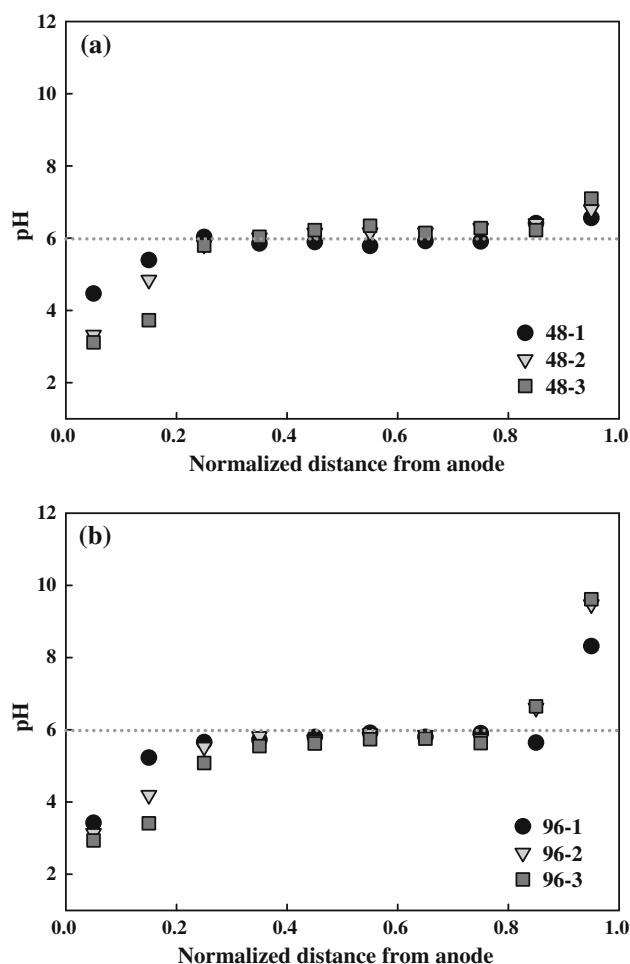
3 Results and discussion

3.1 Soil pH

Electrolysis of tap water, the electrolyte, under the applied voltage caused a change in pH. These electrolysis reactions can be described as



Hydrogen and hydroxide ions generated at the anode and cathode, respectively, were transported toward the opposite charged electrodes by electromigration. Thus, soil pH was decreased near the anode and increased near the cathode. Figure 2 shows the distribution of soil pH after the EK experiment. After 48 h (Fig. 2a), the pH at the middle of the soil compartment was not changed, but small variations were found near the electrodes. The largest change was observed at the 3 V/cm, with the pH ranging from 3 to 7. Compared to the 48 h study, soil pH at 96 h duration was found to be more acidic and basic near the anode and cathode, respectively (Fig. 2b). The increased voltage gradient and electrical input duration lead to a significantly greater electrolysis reaction. A greater amount of hydrogen ion moved toward the soil section by electromigration because its ionic mobility is higher than hydroxide. These

**Fig. 2** Soil pH after EK treatment. **a** 48 h, **b** 96 h

results may provide an explanation for the soil pH being markedly altered at only the closest section from the cathode, whereas pH was decreased at the two sections from the anode.

3.2 Salt concentration in soil compartment

The salts exist as a form adsorbed on soil surface or charged ion in pore water. Adsorbed salts are easily soluble or exchangeable in water and dissolved salts can be transferred by electric fields. Figures 3 and 4 show the distribution of salts after the EK experiment at 48 and 96 h, respectively. Ionic mobility of some salts is shown in Table 3 [30].

3.2.1 Chloride

Chloride (Cl^-) was accumulated above the initial concentration at the two sections from anode and had the highest value (C/C_0) nearest the anode during 48 h (Fig. 3a). The voltage gradient of 3 V/cm showed lower chloride concentration compared to the 1 and 2 V/cm because the electromigration is proportional to the voltage gradient. Chloride was completely removed from only three sections at 1 V/cm, and it was not detected in seven different sections at 2 and 3 V/cm. At 2 V/cm, the chloride concentration in the first and second soil sections from the anode was lower than that at 1 V/cm. This is mainly because at the higher voltage gradients a greater amount of chloride was transferred toward the anode by electromigration, and the

Fig. 3 Removal efficiency (C/C_0) using electrokinetic process during 48 h. **a** Chloride, **b** nitrate, **c** sulfate, **d** potassium, **e** calcium, **f** sodium

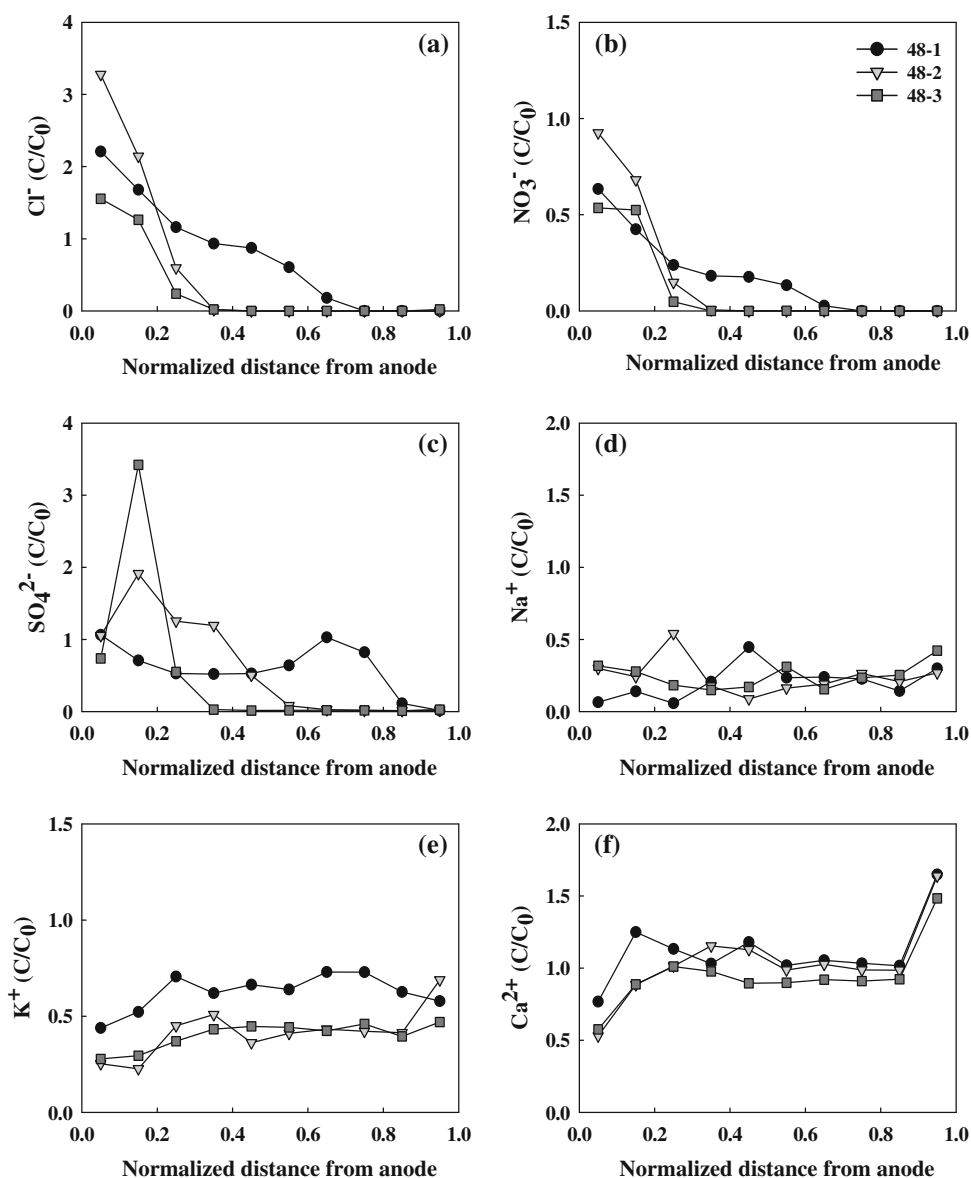


Fig. 4 Removal efficiency (C/C_0) using electrokinetic process during 96 h. **a** Chloride, **b** nitrate, **c** sulfate, **d** potassium, **e** calcium, **f** sodium

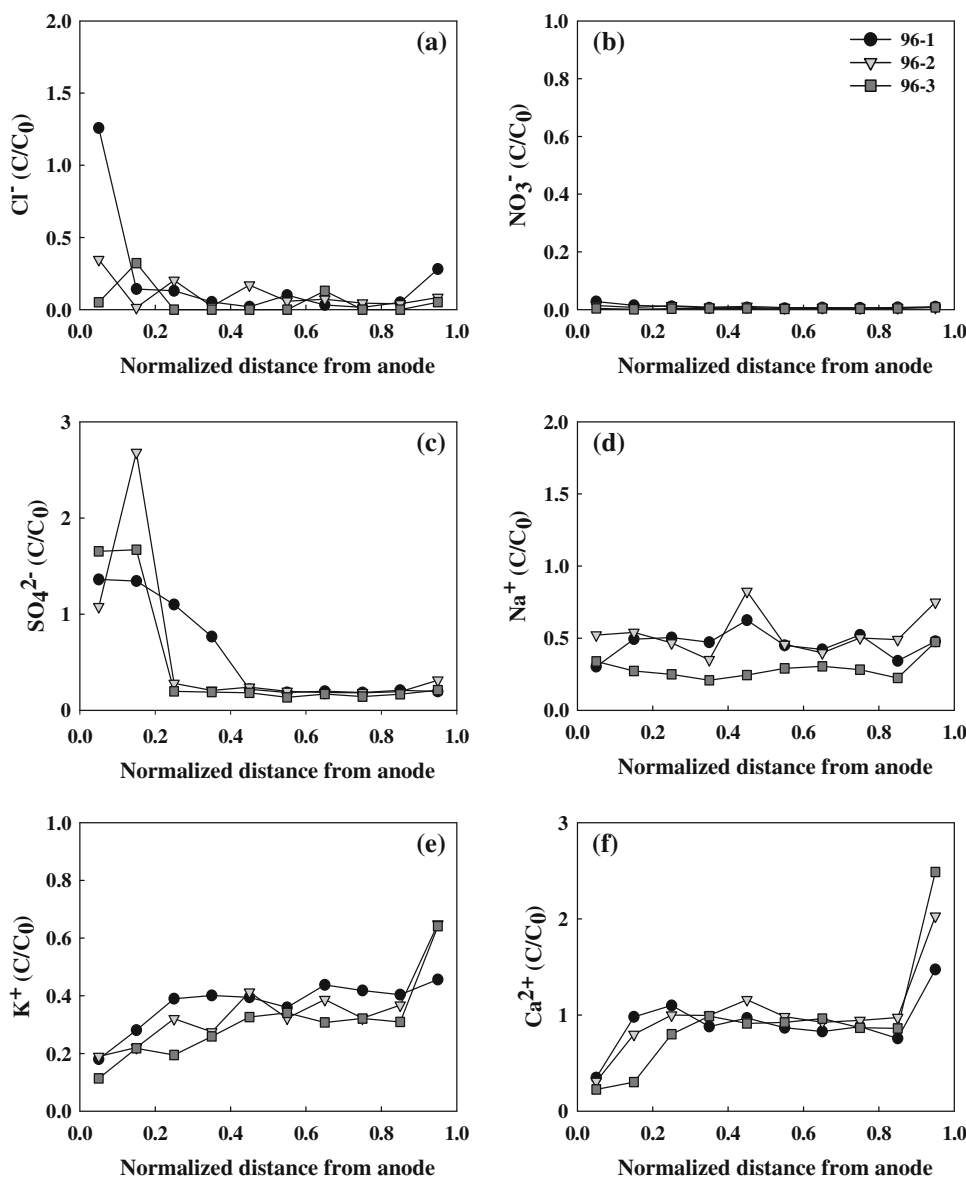


Table 3 Self-diffusion coefficients (D_j), and ionic mobility (u_j) of some salts

Species	$D_j \times 10^6 \text{ cm}^2/\text{s}$	$u_j \times 10^6 \text{ cm}^2/\text{V s}$
Cl^-	20.3	791.7
NO_3^-	19.0	741
SO_4^{2-}	10.6	826.8
Ca^{2+}	7.92	617.8
K^+	19.6	764.4
Na^+	13.3	518.7

amount of accumulation was increased. Chloride accumulated near the anode was removed at 3 V/cm. It appears that chloride diffused and migrated toward the tap water used as anolyte; anolyte concentrations of 2 and 3 V/cm were 47.3 and 67.4 mg/L, respectively. In addition, some chloride can

be converted into chlorine gas (Cl_2) near anode [31], thus the accumulated Cl^- was removed at this section. The chloride removal efficiencies of each voltage gradient were increased in order: 3 (69.1%) > 2 (39.7%) > 1 V/cm (23.6%).

Chloride was removed by greater than 60% from most soil sections except the first section at 1 V/cm and it was removed in all voltage gradients (>80%) after 96 h to a greater extent than at 3 V/cm during 48 h, which had the highest efficiency (Fig. 4a). Approximately 90 and 95% of chloride were removed at 2 and 3 V/cm.

3.2.2 Nitrate

The electromigration was dominated by nitrate removal even in the presence of hydraulic flow toward the cathode

[12, 13, 26, 27]. A low pH can result in the transformation of nitrate into N_2 gas [12]. As a result, nitrate was lower than initial concentration in all soil sections and showed the highest removal efficiency among the other salts. Over 80% of nitrate was removed for all the tests during 48 h (Fig. 3) and little nitrate was still residual in soil compartment after 96 h (Fig. 4).

After 48 h, the entire nitrate were removed from seven sections at 2 and 3 V/cm, while three sections from cathode showed 100% efficiency and accumulated near the anode at 1 V/cm. At the nearest soil section from the anode, nitrate concentration at 2 V/cm was higher than that at 1 V/cm because a greater amount of nitrate was transferred from the middle soil section to the anode by electromigration. At 96 h, nitrate removal was above 99% and little nitrate was observed in the first section from the anode (Fig. 4b). The nitrate concentration at the nearest anode section was decreased as the voltage gradient was increased, due to the greater number of conversions of nitrate to N_2 and diffusions. Removal rate at 48 and 96 h was 23–25 and 13.9–14 mg/h, respectively. The rate was decreased at a longer operation time suggesting that a greater quantity of nitrate was removed at an early stage. A similar pattern was observed for all other salts. Nitrate was removed at the fastest rate among other salts and can be removed within a short time period using electrokinetics.

3.2.3 Sulfate

Sulfate has the highest ionic mobility, however, it was found to have lower removal efficiency than chloride and nitrate. Sulfate was not readily extracted from the soil because it is a divalent and the calcium sulfate complex has very low solubility. The higher the voltage gradient that was applied, the greater the sulfate transfer toward anode occurred by electromigration. The average sulfate removal at 1 V/cm was 40.3%, and it was found to be similar with 2 V/cm (39.3%) during 48 h. At a voltage gradient of 3 V/cm, a gradual accumulation was observed between the second and fourth sections from the anode with most of the sulfate existing at the second soil section. Given that sulfate was only migrated in soil up to 2 V/cm, approximately 51.6% of sulfate was removed from soil at 3 V/cm of 48 h experiment. Sulfate at the middle section of soil in 48 h duration was transferred to anode and accumulated during 96 h. The efficiency in 96 h increased 1–5% compared with 48 h; therefore, most sulfate migrated in the soil section without substantial removal despite an increase in electrical input period.

3.2.4 Calcium

Calcium decreased near anode and accumulated near cathode (Figs. 3, 4). The acidic zone was generated near

anode because of an electrolysis reaction and it is possible that calcium desorption from soil surface was enhanced in the vicinity of the anode. Desorbed calcium's in pore water were migrated and accumulated near cathode. Compared with 48 h duration, 96 h experiment revealed that more calcium was reduced near anode and increased at the nearest cathode compartment up to 2.5 times. However, most of the calcium ions remained in the soil section since calcium was precipitated and strongly adsorbed on soil surface. Also, migrated calcium formed $Ca(OH)_2$ precipitate adjacent to the cathode, and hence calcium was not detected in the EOF solution. Due to these reasons, the removal was only 9%, even though the highest voltage gradient was applied.

3.2.5 Potassium

As shown in Table 3, potassium had the highest ionic mobility among the other cations and the removal rate at 3 V/cm was 21 and 11 mg/h at 48 and 96 h, respectively. Among all the ions, it had the second highest removal rate, with nitrate being the first. In all soil sections, potassium was lower than the initial concentration, and showed a little migration toward the cathode. The 3 V/cm showed the highest efficiency, approximately 60%, and 1 V/cm showed 37.5% removal after 48 h. In case of the 96 h duration, potassium also showed a concentration gradient in the soil that was decreased in anode and increased near cathode compared with the 48 h experiment.

3.2.6 Sodium

Sodium was removed irregularly among all of the soil sections due to its low ionic mobility (Table 3). Sodium concentration at the 96 h was greater than that at 48 h even though electrokinetic treatment time was increased. At anolyte, calcium concentration was unchanged between 48 and 96 h and potassium was increased over time, while sodium was reduced. It appears that diffused sodium in the anolyte migrated toward the soil compartment during 96 h.

3.3 Electrical current and electro-osmotic flow (EOF)

To evaluate the mobility and removal potential of salts under electric conditions, a constant voltage was applied and the electrical current change during experiment was shown in Fig. 5. At the beginning, the currents were under 3.5 mA/cm^2 in all the 48 and 96 h experiments. Next, the current was gradually increased over time indicating that the resistance was decreased by Ohm's law and the electric conductivity was increased [32]. Dissolved salts are

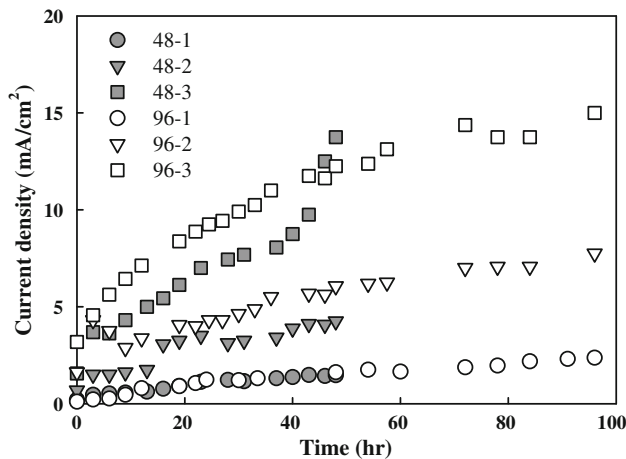


Fig. 5 Electrical current change during EK treatment

transferred and removed by electromigration and electro-osmosis. Removed salts were contained in electrolyte or converted to other chemicals suggesting that current was increased by existing ions in the electrolyte. The average current densities at 1, 2 and 3 V/cm were 1.0, 2.9, and 7.1 mA/cm² for 48 h, 1.3, 5.1, and 10.1 mA/cm² at 96 h duration, respectively. There was a greater increment in current between 2 and 3 V/cm than 1 and 2 V/cm. These findings show that the increasing rate in the electrical current density was faster in high voltage gradient that may be related to removal of salts.

Helmholts (1879) and Smoluchowski (1914) advanced a theory on electro-osmosis that has been widely used. According to this theory, the electro-osmotic flow rate can be estimated by the following [30].

$$Q = k_e \times i_e \times A$$

Q is the electro-osmotic flow rate (m³/s), k_e the coefficient of electro-osmotic hydraulic conductivity (m²/V s), i_e the electrical potential gradient (V/m), and A the total cross-section area (m²)

According to this equation, electro-osmotic flow is affected by the electric gradient between electrodes. Figure 6 shows the accumulated electro-osmotic flow during 48 and 96 h. The amount of EOF was proportional to the voltage gradient, and total volume of 1, 2, and 3 V/cm was 33, 80, and 92 mL at 48 h and 96, 122, and 156 mL at 96 h, respectively.

Anions concentrations in overflowed EOF were decreased as the time elapsed, whereas that of cations was increased. These findings appear to be related to electromigration. For anions, electromigration and EOF were generated in the opposite direction. Therefore, the anions in EOF which was toward the cathode were decreased as time elapsed.

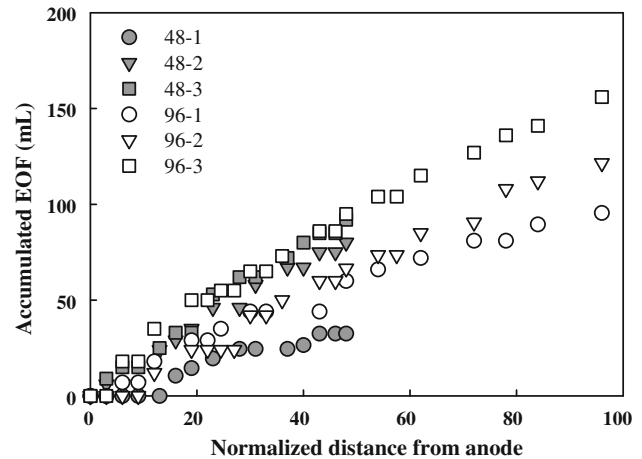


Fig. 6 Accumulated volume of EOF

3.4 EC and energy consumption (kWh/t)

Ions in pore water or at soil surface are influenced directly on a soil EC (dS/m). Soil EC is increased in proportion to the number of ions, namely, high ionic concentration leads to high EC.

Figure 7 shows a soil EC distribution following an EK experiment. EC was found to be higher than initial value near anode at 48 h duration, but it was lower than the initial value in all soil sections at the 96 h experiment. During 96 h, the soil section at the nearest cathode had the high EC value compared with 48 h because more cations such as calcium and potassium were accumulated at high concentration near the cathode by electromigration. Most of the ions were observed near anode in both durations, thus the major existing ions in soil compartment were anions. At the beginning, soil EC was dominated by nitrate due to its high initial concentration, however, this was dependent on sulfate and other salts after electrokinetic process. Since, nitrate can be easily removed from soil EC was less affected by nitrate. EC was analyzed using distilled water, and cations used the NH₄OAc to extract the exchangeable cations. These results suggest that EC shows the only soluble ions to be dissolved by water and EC was not greatly affected by cations although initial concentration of potassium was high.

Most plants are not affected by EC at a range of 1–2 dS/m, and sensitive plants to salts are stressed at 2–4 dS/m. During 48 h, the number of soil sections which had lower value than 2 dS/m was significantly increased with application of the higher voltage. As time elapsed, EC at the middle soil compartment was decreased under guide in 1 V/cm experiment. The lowest EC values for 48 and 96 h were 0.6–0.7 and 1.4 dS/m, respectively; the observed increase during 96 h was mainly due to sulfate and sodium.

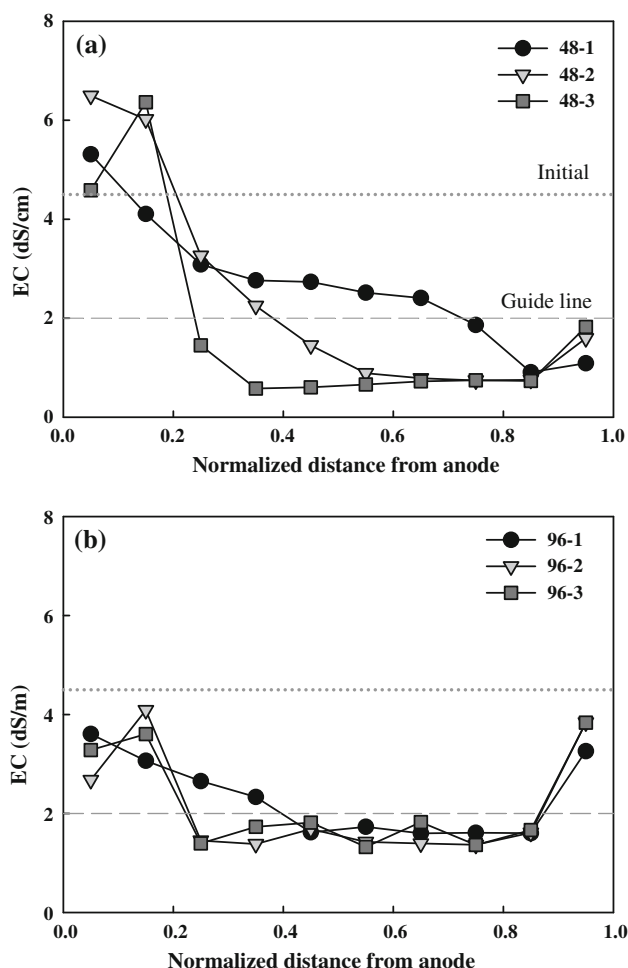


Fig. 7 Soil EC distribution. a 48 h, b 96 h

Table 4 summarized the average soil EC after EK treatment and EC decrease. The guide value (2 dS/m) was achieved at 3 V/cm of 48 h duration (1.8 dS/m). This value was lower than 96 h experiment due to an increase in some salts (sulfate, sodium), extraction of calcium ions, and a high EC value near cathode at the longer durations. The electric energy consumption was high because the saline soil used in this experiment had a high electric conductivity. The voltage gradient of 3 V/cm during 48 h showed the lowest EC value, although energy consumption was

Table 4 Average soil EC (dS/m) after experiment, EC decrement (%) and total energy consumption (kWh/ton) for EK treatment

	48 h			96 h		
	1 V/cm	2 V/cm	3 V/cm	1 V/cm	2 V/cm	3 V/cm
EC	2.7	2.4	1.8	2.3	2.1	2.2
EC decrement	40.4	46.1	59.3	48.7	53.5	51.4
Energy	27.8	161	571	82	626.7	1882.4

very high. Greater amount of energy was consumed at 2 and 3 V/cm, it was not nevertheless significantly influenced on the decrease in EC.

4 Conclusions

An EK process was effectively utilized to reclaim saline soil. Electrical current was large in soil with high EC due to high salinity. Current up to 15.6 mA/cm² under a gradient of 3 V/cm was recorded during a 96 h experiment. Nitrate and chloride were transported and removed rapidly because of their high solubility and conversion to N₂ and Cl₂ gasses near the anode, respectively. Sulfate, which had the highest ionic mobility, had the lowest removal efficiency among the anions. For sulfate and calcium, the removal efficiency did not change significantly between 48 and 96 h. These ions were not easily extracted from the soil and precipitated as calcium sulfate. Potassium concentrations reduced in all soil sections, with remarkable electromigration occurring between 48 and 96 h. A large amount of nitrate, which had the highest initial concentration among anions, was removed from the soil. However, 47–61% of the sulfate in the soil remained after EK treatment, suggesting that soil EC was affected by sulfate ions. Additionally, the EC distribution was comparable with that of sulfate and calcium after 96 h due to calcium accumulation in the vicinity of the cathode. Only the experiment using a gradient of 3 V/cm for 48 h achieved the guideline EC value; however, it was least economical in terms of energy consumption. Further studies are required to remove the sulfate and sodium in an effective manner. EK treatment for extended durations with a low voltage gradient is necessary because soil characteristics and properties can be affected after EK treatment under high voltage gradient and a lot of electricity is consumed.

Acknowledgment This work was supported by a grant from Korea Electrotechnology Research Institute.

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